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Photorefractive and Photochromic Effects in Barium Titanate

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1.0 Introduction

Nominally undoped p-type BaTiO_3 was grown that exhibits interesting intensity dependent photorefractive and photochromic properties. When its photochromism is activated the absorption increases at the activating wavelength and there is a concurrent and proportional reduction in photorefractive response time. Three photoactivated and thermally reversible absorption conditions and the difference spectra of the photochromic crystal are shown in Figure 1.

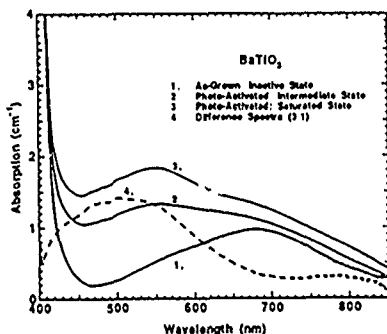


Figure 1.

2.0 Experiments

The photochromism is activated with high intensity ($\sim 3.5 \text{ W/cm}^2$) argon ion laser illumination ($\lambda=514.5 \text{ nm}$). A concomitant change in the crystals' light-induced absorption/transparency properties also originate with the photochromic change. In our photorefractive experimental setup we are able to measure light-induced changes in the absorption by shuttering the incoherent pump beam. Shown in Figure 2 are three signal transmissions corresponding to the three

absorption states shown in Figure 1 and for various pump and signal beam conditions.

Initially, only the signal transmission is observed. Then the pump beam shutter is opened while the pump beam is being vibrated over several grating periods at a rate much greater than the grating formation time (incoherent pump) and thus absorption effects are now observed. When the crystal is in the inactive photochromic state light-induced absorption is observed, in the saturated state it shows light-induced transparency and little or no light-induced absorption is observed in the intermediate state since it is a combination of the inactive and saturated state.

These photochromic states were also examined by measuring the effective gain which is the ratio of the steady-state transmitted signal intensity when both the signal and pump beams are coherently present within the crystal

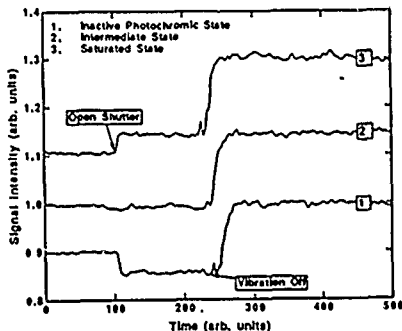


Figure 2.

to the signal intensity transmitted when the pump beam is incoherent with the signal. With this technique, the pump beam continuously illuminates the crystal and the temperature and the absorption are constant in both measurement conditions (coherent versus



incoherent). The measurements were made at 500 mW/cm^2 , low enough to preserve the initial photochromic state. In addition, to determine possible contributions from both electrooptic and absorptive beam coupling [1] we determined the gain for the $\pm C$ -axis orientations of the crystal where the effective gain is given by,

$$\gamma_0^{\pm} \equiv \frac{I_c(L)}{I_{inc}(L)} = \frac{I_s \text{ with Coherent Pump}}{I_s \text{ with Incoherent Pump}} \quad (1)$$

and, for an undepleted pump and small modulation, is equal to,

$$\begin{aligned} \gamma_0^{\pm} &= \exp[\pm \gamma] l / \cos \theta \\ &= \exp[\pm \gamma_{eo} + \gamma_{abs}] l / \cos \theta. \end{aligned} \quad (2)$$

Where, l is the crystal thickness and θ is the internal half-angle between the beams. We note that γ is the gain coefficient which is the sum of the electrooptic gain coefficient, $\pm \gamma_{eo}$, (whose sign is dependent on the orientation of the C-axis and sign of the majority charge carriers), and γ_{abs} , the absorptive "gain" coefficient from trap gratings [1]. To reduce errors from physically reorienting the crystal we simply exchange the roles of the pump and signal beams by changing their relative intensities with the crystal in a fixed position,

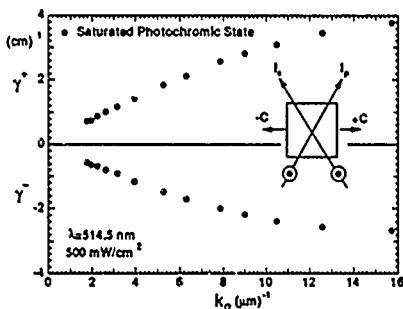


Figure 3. Beam coupling dependence on grating wavevector and c-axis orientation.

(where their beam ratio is 800). Shown in Figure 3 is the gain as a function of grating wavevector for $\pm C$ -axis crystallographic orientations. There is an asymmetry in the beam coupling gain, i.e. $+\gamma \neq -\gamma$, that cannot be attributed to light-induced absorption effects as described by Motes and Kim [2] but could be ascribed to absorptive coupling previously described by A. V. Knyaz'kov and M. N. Lobanov [3] and by Pierce et al. [1].

The intensity dependence of the gain was also determined between 1 mW/cm^2 and 1 W/cm^2 and the sublinear dependence of the response time on intensity was measured over this same interval.

3.0 Theory

In the inactive or saturated photochromic state the crystals absorptive and photorefractive properties are described by the deep and shallow trap model of Tayebati and Mahgerefteh [4] where the absorption due to two levels is,

$$\alpha = s_D N_{DA} + (s_T - s_D) M_0, \quad (3)$$

where s_D and s_T are the excitation cross-sections of the deep and shallow traps, N_{DA} is the difference between the donor and acceptor concentrations and M_0 is the occupied shallow trap concentration. The absorption can be modified by charge transfer from the deep level into the shallow level and depending on its absorption cross-section and concentration it will also modify the absorption characteristics of the material. In the inactive photochromic state light-induced absorption implies that $s_T > s_D$. When a new deep level (2.41 eV) absorbing center is activated to the saturated state light-induced transparency occurs and $s_D > s_T$.

The possible deep and shallow-trap charge gratings, N_1 and M_1 respectively, are,

$$\begin{aligned} -eN_1 &= -mcN_E \frac{k_g^2}{(k_g^2 + k_0^2)} - \\ &mcM_E \frac{1}{(1 + s_T I_0 / \beta)} \frac{k_{0D}^2}{(k_g^2 + k_0^2)} \text{ and,} \end{aligned}$$

$$eM_1 = -meM_E \frac{1}{(1 + \beta / s_T I_0)} \frac{k_g^2}{(k_g^2 + k_0^2)} + meM_E \frac{1}{(1 + s_T I_0 / \beta)} \frac{k_{0D}^2}{(k_g^2 + k_0^2)} \quad (4)$$

I_0 is the intensity, β is the thermal excitation rate of the shallow traps, m is the modulation index, and N_E and M_E are the effective deep and shallow trap densities.

The spatial modulation of the absorption, (giving rise to absorptive coupling), is given by,

$$\hbar\omega(s_D N_1 - s_T M_1), \quad (5)$$

and the space-charge field is intensity dependent where,

$$E_1 = -im \frac{k_B T}{e} \frac{k}{1 + k^2 / k_0^2} \eta(I), \quad (6)$$

where the intensity dependent term has a value between 0 and 1 and is equal to,

$$\eta(I) = \frac{1}{k_0^2} \left[k_{0D}^2 + \frac{k_{0T}^2}{1 + \beta / s_T I_0} \right]. \quad (7)$$

The Debye screening wavevector, k_0 , (which is equal to the square root of the sum of the squares of the deep and shallow trap Debye screening wavevectors, k_{0D} and k_{0T}), is also intensity dependent because of the redistribution of charge between deep and shallow levels.

4.0 Results

From our measurements we are able to determine the electrooptic and absorptive coupling gain coefficients where,

$$\gamma_{\infty} = (\gamma^+ + |\gamma^-|) / 2 \text{ and } \gamma_{\text{abs}} = (\gamma^+ - |\gamma^-|) / 2 \quad (8)$$

Relatively small percentage changes in the electrooptic gain occurred between the three

photochromic states. However, the absorptive gain varied in proportion to the magnitude of the light-induced absorption effects (since they are related through equations 3 and 5). For instance, the intermediate photochromic state, with no light-induced absorption, had virtually no absorptive coupling.

Shown in Figure 4 is a plot of the electrooptic and absorptive gain as a function of grating wavevector with the crystal in the saturated photochromic state. The electrooptic gain approaches zero for small grating wavevectors but the absorptive gain coefficient remained finite as $k_g \rightarrow 0$. (Absorptive coupling was measured at a grating period of 10 μm while no electrooptic coupling was measured. This behavior is predicted by the deep and shallow trap model where the second terms of the charge gratings remain finite at small k_g .)

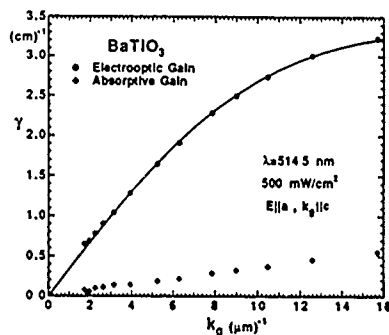


Figure 4. Electrooptic and absorptive gain dependence on grating wavevector.

From the electrooptic gain data of Figure 4 the Debye screening wavevector k_0 and $\eta(I)$ were determined by fitting the equation for the gain as a function of grating wavevector where, $\gamma_{\infty} = (i2\pi\epsilon_{eff} / n\lambda m) E_1$. The relevant electrooptic coefficient is r_{13} and n is the index of refraction. Here, k_0 equals 17.2 μm^{-1} and $\eta(I)$ equals 0.38. The intensity dependence of the space-charge field (eqn. 6), in all photochromic states are nearly identical, where the saturated state gain dependence on intensity is shown in Figure 5.

Measurements of the sublinear dependence of the response time on intensity,

in the saturated photochromic state, give the coefficient of intensity as $x=0.57$. The sublinear dependence of the response time on intensity is predicted by the deep and shallow trap model used here [4] and for a two trap model described by Margerfeth and Feinberg [5] and Brost and Motes [6]. The nominally undoped sample is a type B crystal as described by Margerfeth and Feinberg [5]. The crystal has high dark conductivity with a grating storage time that is intensity dependent and decays in less than a second at a writing intensity of $\sim 3.4 \text{ W/cm}^2$.

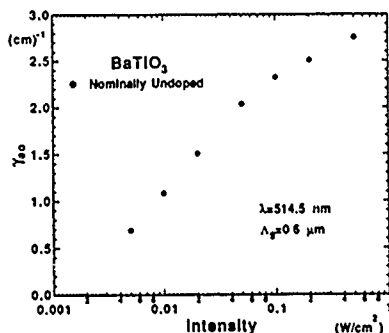


Figure 5. Intensity dependence of the beam coupling gain in the saturated photochromic state.

The response time of the saturated compared to the inactive photochromic state, (or the light-induced decay of the space-charge field), is reduced by a factor of four at 1 W/cm^2 . In one sample the response time was as low as 60 msec at 1 W/cm^2 when the photochromism is saturated (where the $1/e$ intensity decay of the diffracted probe beam was 30 msec). This crystals' sensitivity is believed to be higher than previously reported in the literature at $0.17 \text{ cm}^2/\text{kJ}$.

Similar photochromic and photorefractive changes were observed in LiNbO_3 [7,8]. For those crystals the photochromic behavior was observed only when co-doping the crystal with transition metal ions. However, as was the case for those samples, the changes in the two

visible absorption features of our BaTiO_3 , (at 690 nm and 515 nm), are not proportional implying the interaction of more than two species. When we reduce these crystals, ($P_{\text{O}_2} < 10^{-15} \text{ atm}$), the photochromism is eliminated and thus we suspect that co-doping and oxygen play a role in the photochromism observed here. Mass spectroscopy is being conducted to determine the impurity ions in our samples.

In conclusion, we have reported the photochromic properties of BaTiO_3 that lead to an increase in the absorption and to a proportional decrease in response time. We find light-induced absorption/transparency in the same crystal depending on the photochromic state. The electrooptic and absorptive beam coupling effects were resolved by our measurement technique and characterized in terms of a deep and shallow trap model. In particular, the absorptive coupling is finite for small k_z and for this photochromic crystal when no light-induced absorption changes are observed also no absorptive coupling is observed (and vice-versa).

Acknowledgment

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